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The Electrical Conductivity of Plasma-sprayed
Alumina and Spinel Irradiated at 400 & 500C

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10th February 1995

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Contents

ABSTRACT

The effects of radiation on electrical conductivity in ceramic insulators are briefly reviewed with particular emphasis on the radiation-induced electrical degradation (RIED) effect in which large increases in the intrinsic electrical conductivity have been observed following irradiation within a restricted temperature range whilst subject to electric fields >600V/cm. The aim of the present work was to extend the studies of the RIED effect to plasma-sprayed forms of alumina and spinel.

The experimental arrangement using the Harwell Tandem accelerator is described along with the target which is capable of operating at temperatures up to 900C in a vacuum of $<10^{-6}$mbar. Irradiations were performed on plasma-sprayed alumina and spinel at 400 and 500C using 10MeV protons to damage doses of $>10^{-3}$ displacements per atom (dpa). An electric field of 1kV/cm was applied across the samples during irradiation. The irradiations were interrupted at intervals and the conductivity measured at the irradiation temperature. At the end of the irradiations the temperature dependence of the electrical conductivity was measured. The experimental results showed that for alumina the conductivity decreased for both irradiation temperatures. At the highest damage dose of $4x10^{-3}$ dpa the intrinsic conductivity for irradiation at 400C had decreased by a factor of ~3 and for the 500C irradiation by a factor of ~10. The conductivity of spinel irradiated at 400C increased by factor of ~3 whilst that irradiated at 500C showed only a slight increase.

This report covers both interim and final reports.

1. INTRODUCTION

Hodgson [1,2,3,4,5] has shown that when alumina is simultaneously exposed to a moderate electric field, moderate temperature and radiation damage, the intrinsic conductivity may increase by many orders of magnitude. This has been termed radiation-induced electrical degradation (RIED). The electric field may be ac or dc [5] with a major threshold occurring at ~ 600V/cm. Degradation still occurs with lower electric fields but after a much longer incubation time. Both ionisation and displacement damage appear to be necessary features [1] and RIED has been observed within the temperature range 200-550C. The effect occurs at lower temperatures but over longer time scales. Hodgson used 1.8MeV electrons but the effect has also been seen with ion and neutron irradiations. Pells [6] irradiated alumina with 18MeV protons at 400 and 500C with an applied electric field of 5kV/cm. Permanent increases in the conductivity occurred for the 500C irradiation only, although the increase was very large giving a conductivity of ~10^{-3}S/m after a damage dose of 2.5 x 10^{-3}dpa. Similar changes were reported by Ivanov et al [7] for neutron irradiations at 445C. More recently Moslang et al [8] has shown that different grades of alumina may give widely differing results, ranging from no RIED effect to very large effects.

The onset of degradation occurs after ~10^{-5}dpa for both electron and proton irradiations and has saturated after ~10^{-2}dpa. These are very low doses in radiation damage terms. There is no generally accepted model for the RIED effect but it appears to be associated with the formation of a second phase [3] which can be of sufficient size to be seen with an optical microscope. Pells [9] has shown that, in the case of electron irradiated sapphire, the second phase is gamma alumina. The temperature range over which RIED occurs suggests that diffusion on the oxygen sublattice is involved as aluminium Frenkel pairs are practically immobile below 400C. This is supported to some extent by the observation [4] that there is an enhanced production of F^+ centres (an oxygen vacancy with one trapped electron) during irradiation with an applied electric field, although how this leads to nucleation of gamma alumina after a dose of 10^{-5}dpa remains unclear.

The original aim of the present work was to determine whether there was a measurable RIED effect in plasma-sprayed alumina and spinel irradiated with 10MeV protons at temperatures of 400C and 700C while subject to an electric field of 1kV/cm. The requirement to irradiate at 700C was subsequently modified to 500C in the light of previous measurements on sapphire irradiated at temperatures above 600C whoch showed that the electrical conductivity decreased with increasing damage dose.

In order to present a coherent picture of the work no separate interim report has been prepared on the 400C irradiations alone and this one report combines both interim and final reports.

2. EXPERIMENTAL METHODS

2.1 The Irradiation Facility

The target assembly was built on the end of one of the beam lines attached to the Tandem accelerator equipped with full beam steering and focusing lenses and operating at a base vacuum pressure of $< 1 \times 10^{-6}$mbar. The Tandem accelerator has a maximum proton beam energy of 12MeV and a beam current of ~ 5uA. The target assembly consisted of two major sections. The first was a collimator box pumped by a 280l/s diffusion pump. The collimator consisted of four air cooled quadrants which defined a 10mm diameter aperture. The beam current on each quadrant could be monitored individually. A water cooled beam stop could be inserted immediately after the quadrants and was used for setting up the irradiation beam. The quadrant box was separated from the target chamber by a 300mm long, 25mm diameter tube to provide a measure of differential pumping between the two sections.

The specimen chamber was 450mm long by 150mm diameter with five major ports plus two small ports for vacuum gauges and two angled ports for observation of the target area. A second set of quadrants was mounted at the beam entry port. The chamber was pumped by a 1000l/s cryopump giving $\sim 10^{-8}$mbar base pressure and $< 10^{-7}$mbar during proton irradiation with the sample at 700C. The conductivity target was mounted on the end port and the whole of the target chamber was insulated from earth so that the target sample could be used as a Faraday cup to measure beam current. In practise irradiation with an electric field applied to the sample made it impossible to measure beam current directly and the current had to be measured by periodically inserting the beam stop into the proton beam during the course of an irradiation. The ratio of the target current to the beam stop current was determined before the irradiation started. The 10MeV proton beam was adjusted to give a slightly divergent beam such that a small fraction of the beam was stopped by each of the 8 quadrants. Correct alignment of the proton beam was obtained by ensuring that equal currents were maintained on all of the quadrants. The divergent beam also ensured that the beam flux across the target face did not vary by more than 20%.

2.2 The High Temperature Target

The sample holder had to compromise between being able to dissipate the irradiating beam power at low irradiation temperatures and yet allow the sample to be heated to high temperatures when required. Movement of the sample relative to the beam, and the electrical constraints due to thermal expansion, also had to be minimised. This was achieved by arranging for the sample to be carried on a heater supported inside a nest of stainless steel tubes coaxial with the irradiating beam as shown in Figure 1. The far end of the heater was air cooled to protect the vacuum seals and the whole heater was filled with helium gas. A controlling

chromel-alumel thermocouple ran up the centre of the heater to within 1mm of the sample. On test the heater was run up to 900C and the vacuum pressure in the chamber returned to <10^{-6} mbar after initial outgassing.

The plasma-sprayed alumina and spinel samples, supplied by P. Agnew of TSET, were 150μm thick with niobium centre and guard ring electrodes evaporated on the front face and bonded to niobium discs at the rear. The niobium discs also served to clamp the samples to the sample holder.

Electrical contacts to the centre and guard electrodes of the sample were made by tungsten springs carried by a MACOR glass plate supported by three stainless steel tubes. The three tubes also served as the electrical shields for the leads between the tungsten springs and the leadthroughs on the main vacuum flange which also carried the heater.

2.3 Experimental Procedure

Prior to irradiation the sample was heated to 700C and held at that temperature to outgas the system. The electrical conductivity was also measured during the warming and cooling cycles. Previous work had shown that the conductivity varied with thermal cycling before irradiation, therefore the samples were thermally cycled several times before conductivity measurements were made.

As mentioned before the 10MeV proton beam was set up to give a slightly divergent beam just touching all eight segments of both collimators. A final collimator 9.5mm diameter was positioned 30mm in front of the target sample. This stopped the annular space between the centre electrode and guard ring of the sample from being irradiated and so prevented a stable and possibly conducting layer forming from residual organic gases in the vacuum system and bridging the inner and outer electrodes during the long irradiations.

Once the beam had been set up an electric field of 1kV/cm was applied across the thickness of the sample and the irradiation was started. The change in sample temperature due to beam heating was monitored by an infra-red thermometer and the heater controller adjusted to give the required irradiation temperature. Beam heating produced temperature increases of 20-30C with proton beam powers of 10-30W/cm^2. The beam heating increased during the course of an irradiation as the thermal conductivity of the sample decreased due to radiation damage. This was compensated for by adjusting the set point of the temperature controller and the nominal irradiation temperature was held to $\pm$ 10C throughout an irradiation. The beam current was monitored on the first collimator flap every half hour but rarely varied by more than 10% of the original setting during the course of a day's irradiation.

In order to avoid possible annealing effects the electrical conductivity was

measured only twice at the irradiation temperature during the course of the irradiation. At the end of a days irradiation the sample was allowed to cool with the sample still exposed to the irradiating proton beam and at the beginning of the next days irradiation the proton beam was switched on before the sample was brought up to the operating temperature and the electric field applied. At the end of each irradiation experiment 24 hours was allowed to elapse before any conductivity measurements were made so that the induced activity could decay to a level that could not possibly effect the conductivity measurements. The final conductivity measurements for each sample were made on both heating and cooling cycles with a highest temperature of 700C.

3. EXPERIMENTAL RESULTS

3.1 Plasma-sprayed Alumina

Irradiation at 400C to a damage dose of 4.0×10^{-3}dpa produced a small decrease in conductivity as shown in Fig. 2 where it can be seen that the temperature dependence remains essentially the same, with little change in the activation energy for electrical conductivity at the higher temperatures (see Table 1). Irradiation at 500C produced a larger change, with the conductivity (Fig.3) showing a tenfold decrease after the highest dose of 3.9×10^{-3}dpa.

The variation in conductivity with damage dose for irradiation at both 400C and 500C is shown in the log/log plot of Fig.4. It can be seen that the conductivity begins to decrease after a damage dose of $>1 \times 10^{-4}$dpa.

Table 1 High temperature activation energy of plasma-sprayed alumina and spinel samples 10MeV proton irradiated with an applied electric field of 1kV/cm.

Sample	Irradiation Temperature (C)	Damage Dose ($\times 10^{-3}$dpa)	Activation Energy (eV)
Alumina	400	-	1.15
"	"	3.97	1.14
Alumina	500	-	1.15
"	"	3.90	1.15
Spinel	400	-	1.28
"	"	4.23	1.21
Spinel	500	-	1.37
"	"	3.32	1.31

3.2 Plasma-sprayed Spinel

Irradiation at 400C increased the conductivity by a factor of ~3.5 after a damage dose of 3.5×10^{-4}dpa as shown in Fig.5. At higher doses the conductivity remained fairly constant and may even have fallen slightly at the highest dose of 4.2×10^{-3}dpa. The activation energy for electrical conductivity decreased slightly at the higher temperatures as can be seen in Fig. 6 and is given in Table 1.

Irradiation at 500C again produced an initial increase in conductivity by ~2.5 after a damage dose of 1.05×10^{-3}dpa although the difference had become negligible at the highest dose of 3.3×10^{-3}dpa (see Fig.5). However, the activation energy for electrical conductivity again decreased as shown in Fig.7 and Table 1.

4. DISCUSSION

The results obtained in these irradiations of plasma-sprayed alumina clearly show no indication of RIED as the electrical conductivity decreased for both irradiation temperatures and the activation energy for electrical conductivity remained constant within experimental error. The irradiation temperatures of 400C and 500C are within the range for which Hodgson [1-5] and Pells[6] observed large increases in the intrinsic conductivity when high purity aluminas were irradiated with electric fields of 1.3 and 5.0kV/cm respectively. However, the recent work of Moslang et al using 104MeV helium ion irradiation at a temperature of 450C with an applied electric field of 1kV/cm has shown that very large differences in behaviour can be found in aluminas of different purity. In the case of high purity (99.9%), fine grained Vitox alumina irradiation increased the intrinsic conductivity to 4×10^{-2}S/m after only 1.5×10^{-2}dpa. This is in good agreement with the data of Pells [6]. Wesgo 99.2% alumina irradiated under the same conditions showed a decrease in conductivity similar to that observed in the present work.

In the case of plasma-sprayed spinel irradiated at both 400C and 500C there were small increases in conductivity ranging from factors of 3-4 with a tendency for the conductivity to decrease again at the highest doses. These increases are negligible compared to the large increases observed by Pells [6] for 20MeV proton irradiation of high purity spinel, although the increases in activation energy for conductivity suggest that RIED-like processes may be in operation.

5. CONCLUSIONS

1) The plasma-sprayed alumina samples supplied for this work and irradiated with an applied electric field of 1kV/cm showed significant reductions in conductivity with increased displacement damage dose. Such radiation hardening would be beneficial under most operating conditions.

2) The plasma-spayed spinel showed small increases in conductivity along with

small increases in activation energy after irradiation with an applied electric field which might be consistent with RIED. However, the changes in conductivity would be unlikely to effect the materials operational performance at these damage doses.

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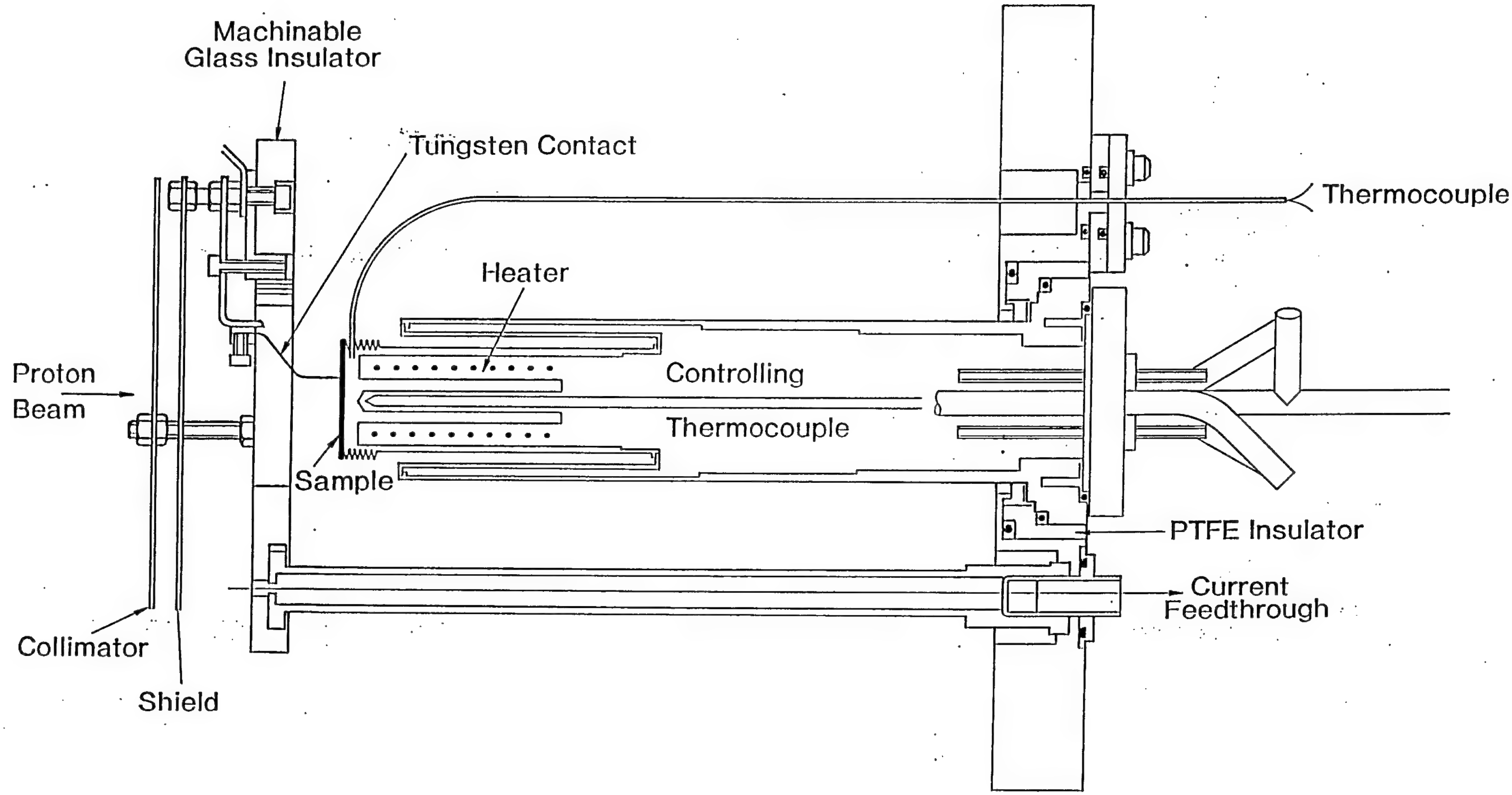

Fig. 1.
Cross section of target assembly

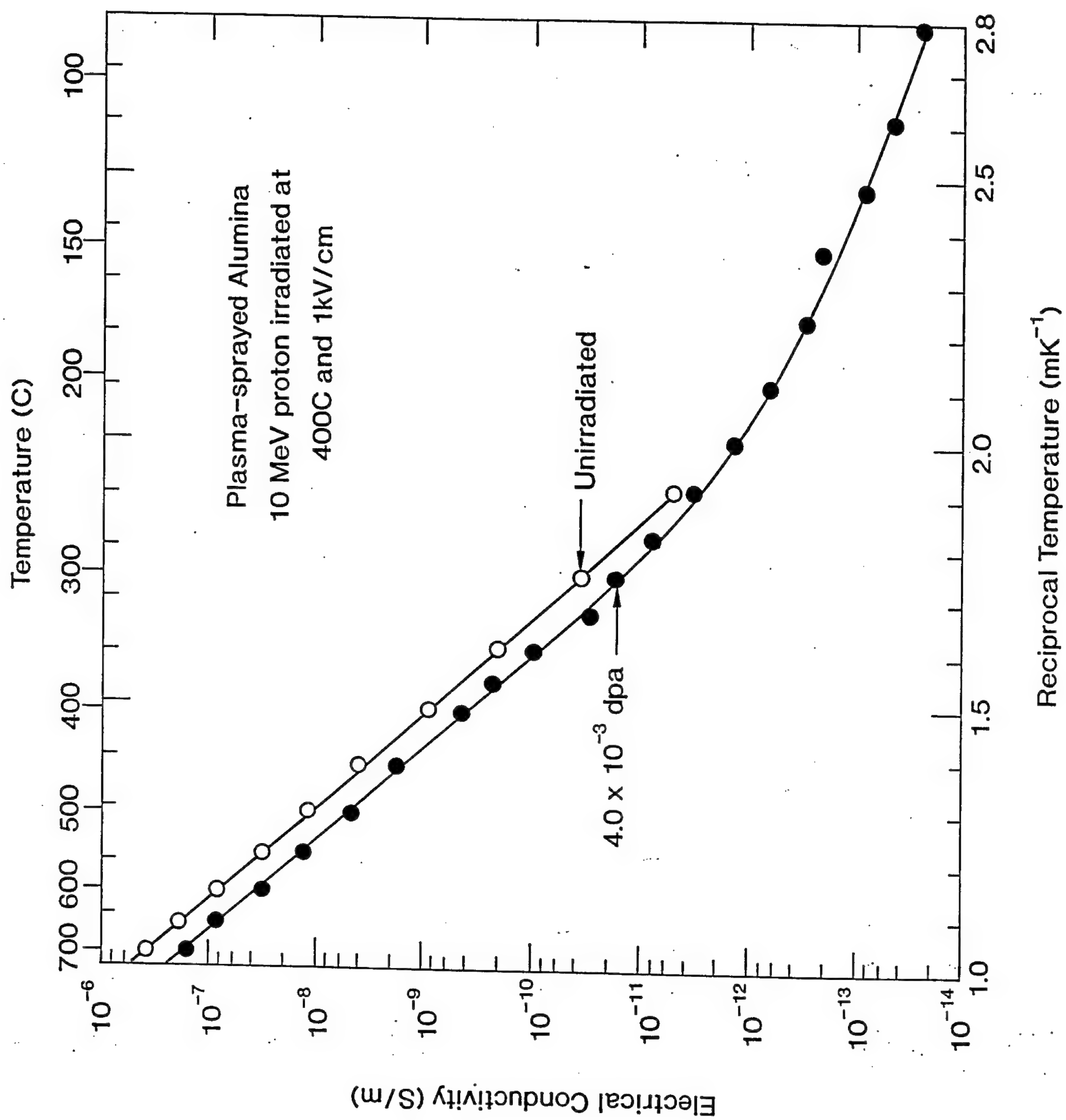

Figure 2.
The intrinsic electrical conductivity of plasma-sprayed alumina as a function of temperature after 10MeV proton irradiation at 400C to the indicated damage doses with an applied electric field of 1kV/cm.

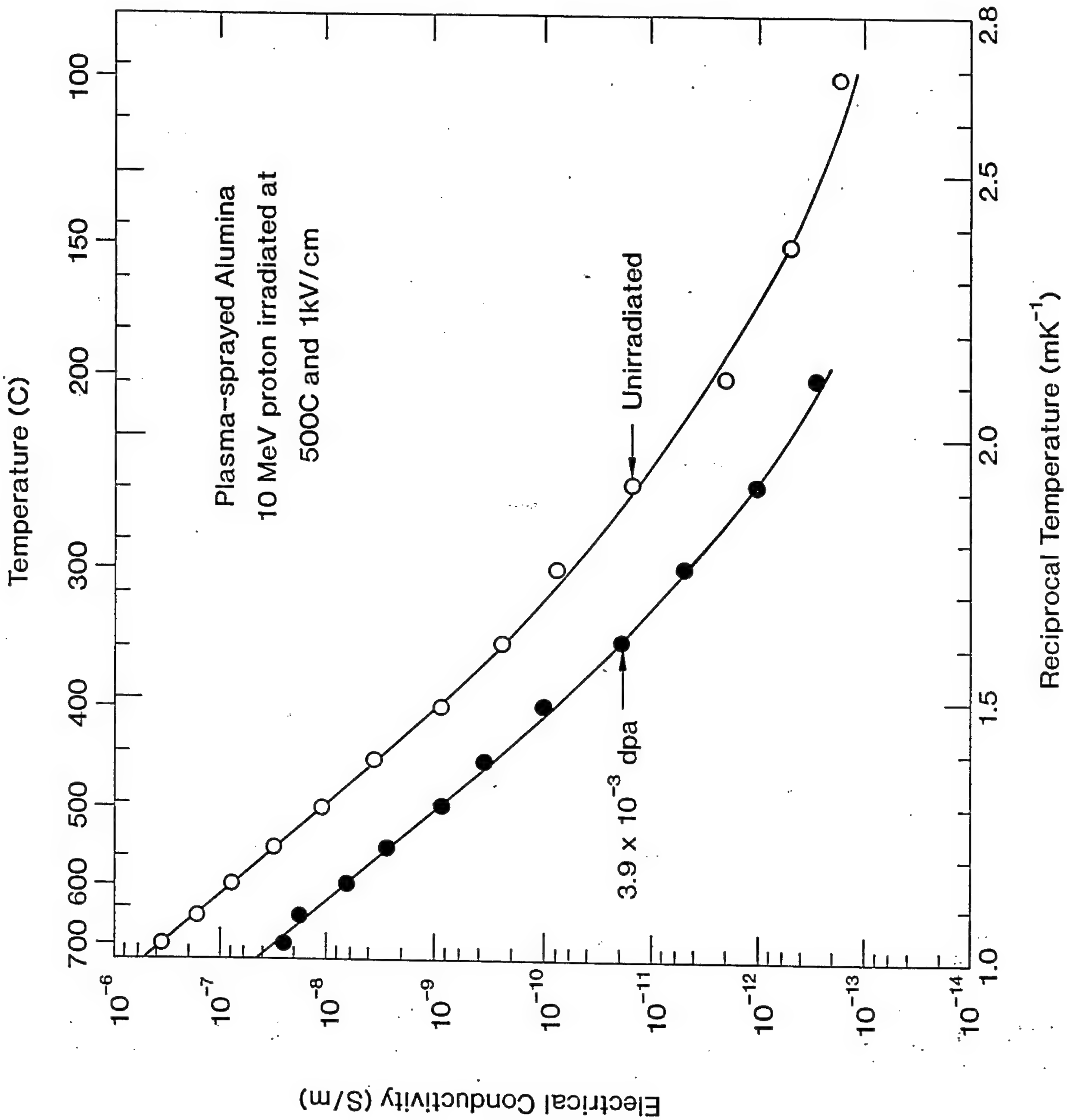

Figure 3
The intrinsic electrical conductivity of plasma sprayed alumina as a function of temperature after 10MeV proton irradiation at 500C to the indicated damage doses with an applied electric field of 1kV/cm.

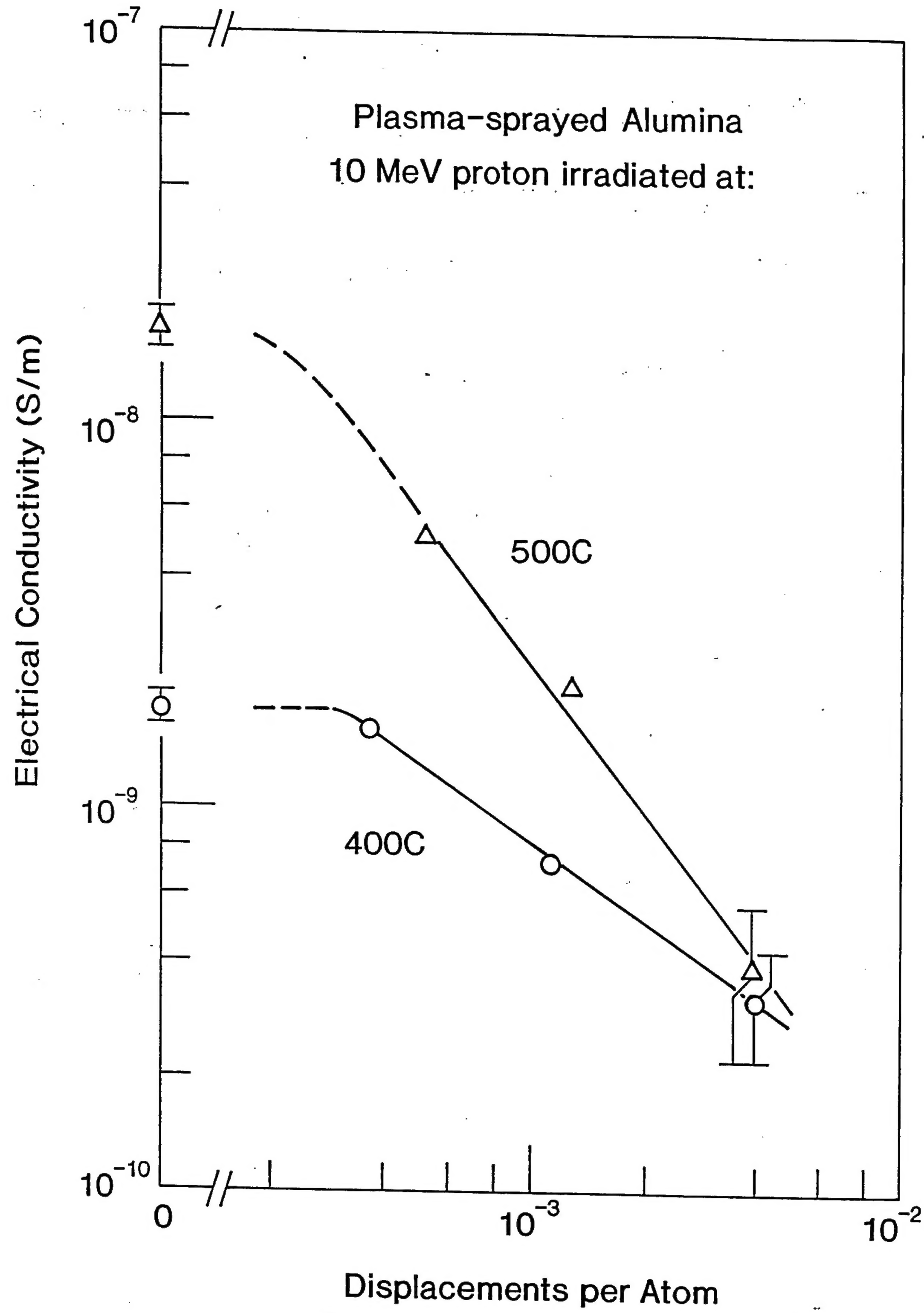

Figure 4.
The intrinsic electrical conductivity of plasma sprayed alumina as a function of damage dose after 10MeV proton irradiation at the indicated temperatures with an applied electric field of 1kV/cm.

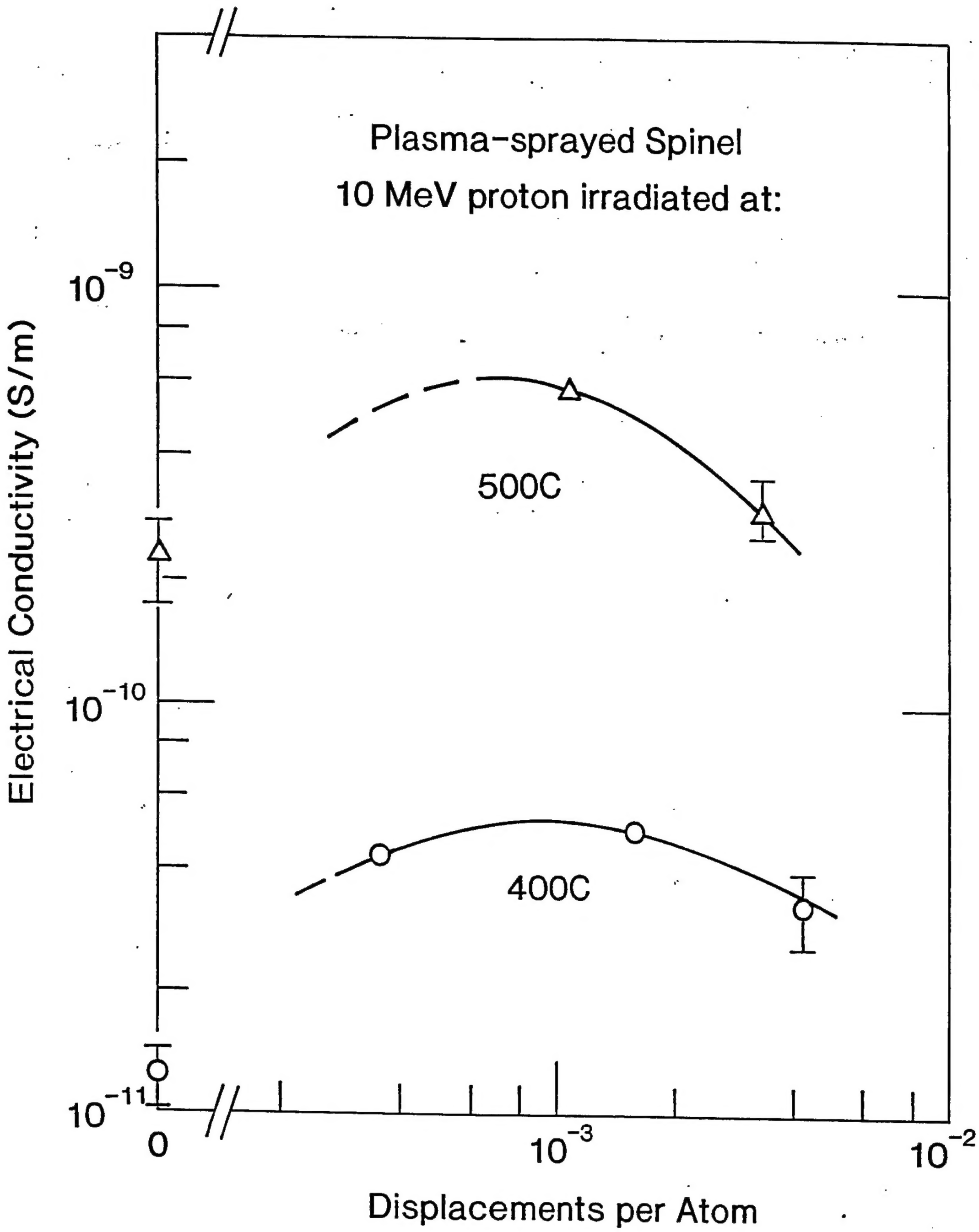

Figure 5
The intrinsic electrical conductivity of plasma-sprayed spinel as a function of damage dose after 10MeV proton irradiation at the indicated temperatures with an applied electric field of 1kV/cm.

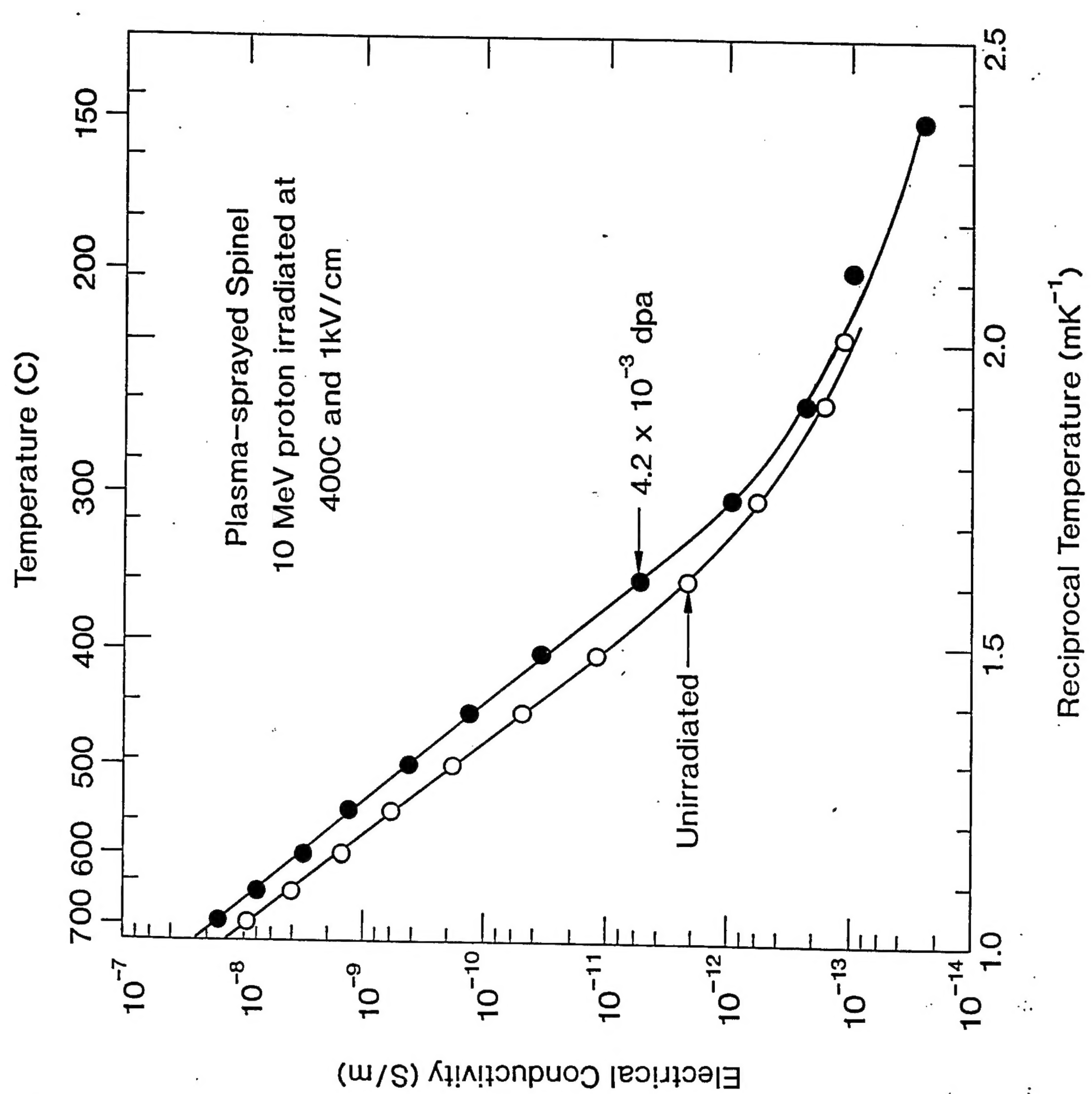

Figure 6.
The intrinsic electrical conductivity of plasma-sprayed spinel as a function of temperature after 10MeV proton irradiation at 400C to the indicated damage doses with an applied electric field of 1kV/cm.

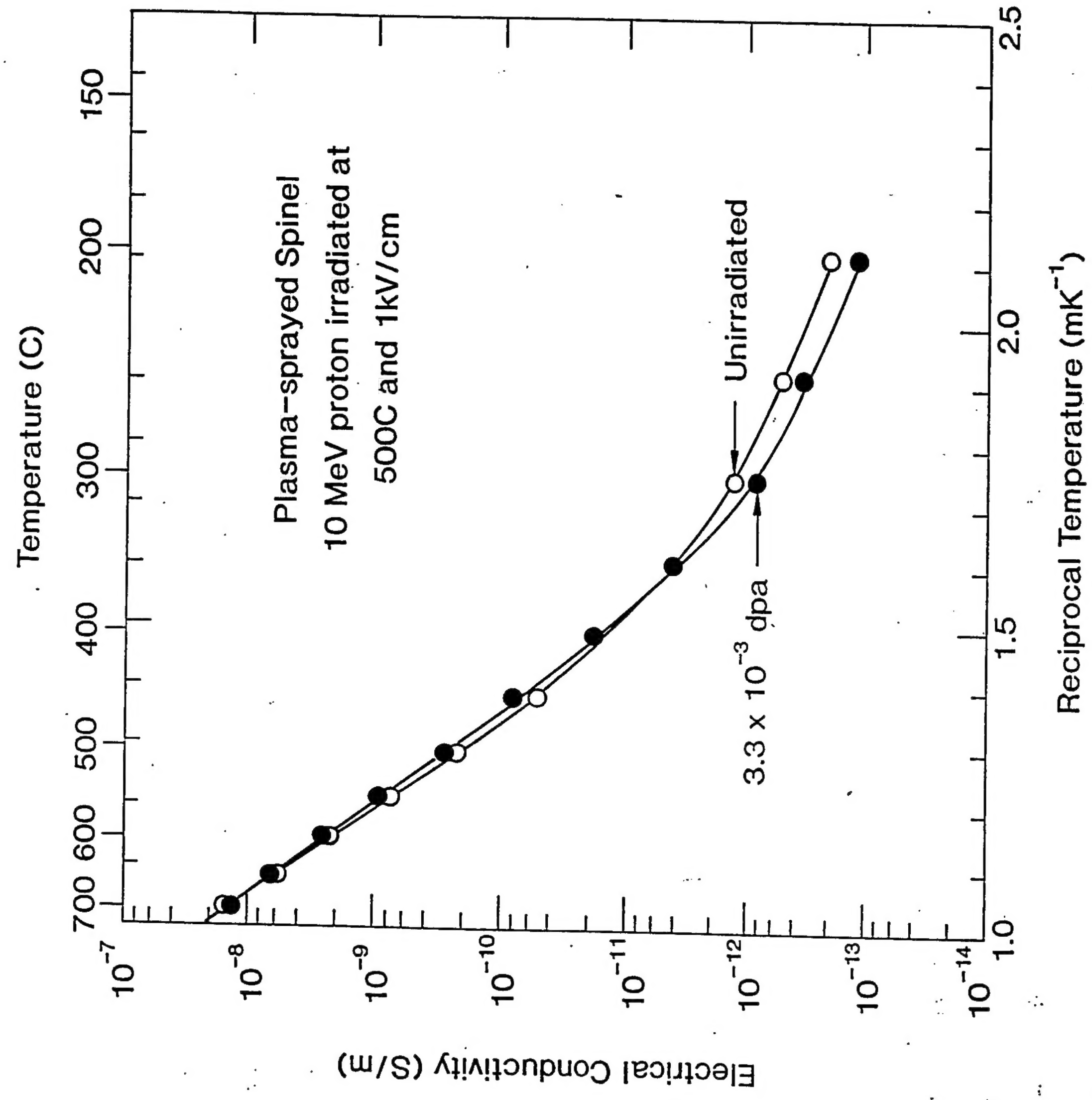

Figure 7.
The intrinsic electrical conductivity of plasma-sprayed spinel as a function of temperature after 10MeV proton irradiation at 500C to the indicated damage doses with an applied electric field of 1kV/cm.